



Lead exposure assessment from study near a lead-acid battery factory in China

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ABSTRACT

The production of lead-acid battery in China covered about one-third of the world total output and there are more than 2000 lead-acid battery factories. They may cause the major environment lead pollution. Blood lead levels of several hundreds of residents were over 100 µg/L due to the waste discharges from a lead-acid battery factory in Heyuan, Guangdong province. This study aimed to find out the environmental lead sources, the human lead exposure pathways, and the amplitudes from a lead-acid battery factory. The study results showed that lead levels in soil, dust, tree leaves and human blood declined with the distances increased from the production site. Twenty nine of 32 participants had blood lead levels of over 100 µg/L with an exceptional high value of 639 µg/L for one child. This result suggested that the lead-acid battery production from this factory has caused the elevated lead levels in its neighboring environment and residents. Dust intake was the dominant exposure pathway for humans (over 90%). The lead levels found in adult and toddler (6.19 and 50.1 µg/kg/d, respectively) in the polluted area were far higher than the provisional tolerable weekly intake (PTWI) of 25 µg/kg body weight (translated into 3.5 µg/kg/d), which was established by the joint FAO/WHO Expert Committee. Blood lead levels within the family members were strongly correlated with the house dust lead levels. Our results in this study suggested that further studies in this area should be performed to assess human exposure and relevant human health risks from living close to lead-acid battery factories.

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1. Introduction

Toxic effects of lead on wildlife and humans may range from enzyme inhibition, anemia to disorders of the nervous, immune and reproductive systems, impaired kidney and cardiovascular functions, and even death with high dosage (IPCS, 1995). These effects can occur over a broad range of exposure. Even low dosages could cause high body burden, and children are more vulnerable than adults to lead exposure because they are in the phase of growth and development and have immature detoxification capabilities. Lead has more profound effect on the brain and nervous system of children. Hand-to-mouth activities and playing pattern may intake more lead (IPCS, 1995; Meyer et al., 2008). The major anthropogenic sources of lead are from fossil fuels, ores, as well as other extracted, treated and recycled minerals. Industrial usage of lead could disperse into environment (UNEP, 2010).

In 2008, the production 90.77 million kilovolt-ampere-hour (kVAh) of lead-acid battery in China counted about one-third of the world total output. To produce these batteries utilized 1.92 million tons of lead, counted as 67% of the total lead usage in China (Zhao et al., 2009). In

China, there are more than 2000 lead-acid battery factories, and 1800 of them are small and medium scale plants without a production permission certificate (<http://news.xinhuanet.com>). These factories failed to meet the national clean production standards, and constituted the major environmental lead pollution. Of the fourteen Chinese pollution incidents in 2010, nine were caused by the lead pollution, and these incidents had resulted in the human blood lead levels within the impacted population over the national standards (100 µg/L). In addition, seven more lead poisoning incidents occurred from January to May in 2011 (<http://news.cntv.cn>). From 2009 to 2011, lead poisoning incidents in several provinces of China had affected more than 4000 children (Ji et al., 2011). High blood lead levels were reported in the working populations within the lead smelter and mining area, as well as in the lead-acid battery factories (Gulson et al., 1994; Trepka et al., 1997; Hsiao et al., 2001; Paoliello et al., 2002; Chuang et al., 2008). However, the effects of lead-acid battery production on the neighboring environment and human blood lead levels of the residents living near the factories were rarely studied/reported. There are about 2000 lead acid battery factories operating in China with unknown environmental hazards. Blood lead levels of several hundreds of residents were over 100 µg/L due to exposure to the waste discharges from a lead-acid battery factory in Heyuan, Guangdong province. Therefore, this study was designed to find out the environmental lead sources, the human lead exposure pathways, and the amplitudes from a lead-acid battery factory.

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2. Materials and methods

2.1. Sample collection

All environmental samples and human blood samples were collected around a lead-acid battery factory in May 2011. The factory, with a battery production capacity of 1.0 million kVAh and usage of 10 thousand tonnes of lead yearly, is located in Heyuan, Guangdong Province. There are a total of 147 environmental samples collected from the factory area, near-by polluted and considered clean-reference areas. These samples include air, water, sediment, soil, tree leaves, dust, food, lead dust and lead powder. All the sampling methodology and information was summarized in Table 1 and Fig. 1. As recommended by the health protection zone criteria for lead-acid battery factories (GB, 11659-89), a health protection zone was set at a distance of 300 m to 800 m around the lead-acid battery factory on the base of average down wind speeds in the last 5 years. Therefore, we defined the zone of 0–500 m distance from the factory as the polluted area and 1000–2500 m as the clean reference area, respectively.

Table 1
Sampling information and lead levels of the collected samples.

Types	The factory area		The polluted area (0–500 m)		The clean area (1–2.5 km)	
	SN ^a	Lead levels	SN	Lead levels	SN	Lead levels
<i>Soil (mg/kg)</i>						
Agricultural soil	0	ND ^b	12	261 ± 582 (50.8–2106)	5	60.5 ± 10.2 (51.0–76.1)
Non-agricultural soil	3	11,318 ± 19,126 (214–33,400)	10	1088 ± 2838 (50.2–9140)	1	60.3
<i>Sediment (mg/kg)</i>						
Pond sediment	0	ND	1	218.4	1	67.75
<i>Water (μg/L)</i>						
Well water	0	ND	8	3.98 ± 3.45 (0.71–9.35)	2	2.88–7.73
Tap water	0	ND	5	1.67 ± 1.68 (0.71–4.59)	1	0.71
Surface water	0	ND	1	5.42	1	3.57
Waste water	1	1240	0	ND	0	ND
Air (μg/m ³)	0	ND	2	0.13–0.24	2	0.10–0.18
<i>Outdoor air</i>						
<i>Food (mg/kg)</i>						
Vegetable	0	ND	14	0.058 ± 0.064 ^c (0.001–4.36)	6	0.041 ± 0.030 (0.012–0.090)
Egg	0	ND	4	0.071 ± 0.024 (0.050–0.102)	1	0.04
Rice	0	ND	4	0.083 ± 0.026 (0.054–0.101)	4	0.077 ± 0.037 (0.048–0.137)
<i>Tree leaf (μg/g)</i>						
Mango	2	107.4–440	2	27.1–29.8	1	4.73
Acacia	1	127.65	1	46.0	0	ND
Banyan	2	36.6–109.1	0	ND	0	ND
Guava	0	ND	1	17.9	1	0.14
Grape	0	ND	1	0.26	1	4.7
Longan	0	ND	1	12.8	1	0.73
Litchi	0	ND	1	23.4	1	4.04
<i>Dust (μg/g)</i>						
Outdoor	2	155,850–351,440	10	8417 ± 6516 (270–19,490)	4	308 ± 16 (290–329)
Indoor	7	659,192 ± 346,287 (13,570–708,560)	11	4741 ± 3694 (506–10,950)	4	331 ± 189 (234–610)

^a SN: sample numbers.

^b ND: not detected.

^c The average lead level of vegetables in polluted area is 4.36 mg/kg with the exception of high lead concentration in water convolvulus vegetable sample.

Fifteen families (age range 1–96 years old) were recruited with 11 of them resided in the polluted area and 4 in the clean-reference area. All the residents grow some of their food from their own field or purchase the rest from the local markets. Rice, eggs and vegetables were collected from their homes. The commonly consumed vegetables in both areas were chive (*Allium schoenoprasum* L.), sweet potato leaves (*Ipomoea batatas*), eggplant (*Solanum melongena* L. egg-plant), green pepper (*Capsicum frutescens* L. green pepper), garlic sprout (*Allium sativum*), water convolvulus (*Herba Ipomoeae Aquaticae*), amaranth (*Amaranthus gangeticus* Linn), and lettuce (*Cichorium endivia*). Drinking water samples, like well water and tap water, were also collected from the recruited families. Tree leaves were also collected from the representative local tree species, like mango tree (*Mangifera indica*), grape (*glucosum*), longan (*Dimocarpus longan* Lour), guava tree (*Psidium guajav*), acacia tree (*Albizzia julibrissn*), banyan tree (*Ficus microcarpa*) and litchi (*Litchi chinensis* Sonn.). Dust samples were collected following a standardized sampling method developed by the Verein Deutscher Ingenieure (VDI, 2001; Huang et al., 2010; Chen et al., 2011). Dust on the surfaces of upholstery, furniture, floor and windowsill was swept using clean paint brushes, and sealed in polyethylene zip bags. During the recruitment, family members were required to provide their personal information of age, body weight, occupations, residential history, dietary habit, smoke habit and health history through filling a questionnaire provided. The present study obtained ethics approval from South China Institute of Environmental Sciences, Ministry of Environmental Protection. All the participants joined this investigation on a voluntary basis.

2.2. Sample preparation and analysis

2.2.1. Soil, indoor dust and sediment

All soil or sediment samples were dried in a freeze drier, ground with mortar and pestle, and then sieved through a 100 μm nylon mesh. About 0.1–0.5 g soil or sediment was digested in 4.0 mL aqua regia overnight in a 100 mL polytetrafluoroethylene (PTFE) beaker, in an electric oven and heated at 190 °C to evaporate the solution volume to about 2.0 mL. A mixture of 5.0 mL of concentrated high-purity HNO₃, 4.0 mL of HF and 2.0 mL of HClO₄ was added and heated. This step was repeated until no perceptible particles existed in the solution. After cooling, the solution was poured into a polyethylene tube, and diluted to 10.0 mL or 25.0 mL with 2% of high-purity HNO₃ solution.

2.2.2. Food and tree leaves

The samples were rinsed with deionized water, dried at room temperature, and homogenized. About 1.5–3.0 g sample was digested with 10.0 mL of HNO₃/HClO₄ mixture (v/v, 9/1) overnight in a 100 mL PTFE beaker and heated at 190 °C. After cooling, the solution was poured into a polyethylene tube, and diluted to 10.0 mL or 25.0 mL with 2% of high-purity HNO₃ solution.

2.2.3. Water

20.0 mL of water was added with 2% of high-purity HNO₃ before sampling. After shipped to the laboratory, water samples were filtered through 0.22 μm filter and lead was instrumentally measured directly.

After sample digestion, lead contents were first determined by flame atomic absorption spectroscopy (FAAS, PerkinElmer AAnalyst 800). However, for those samples with lead contents below the instrument detection limit of FAAS, they were analyzed by graphite furnace atomic absorption spectroscopy (GFAAS, PerkinElmer AAnalyst 800).

2.3. Quality control and quality assurance

Every 10 authentic samples were analyzed as a batch, including a method blank, a duplicate sample and two spiked blank samples. Low lead levels were detected in both food and soil blanks (mean:

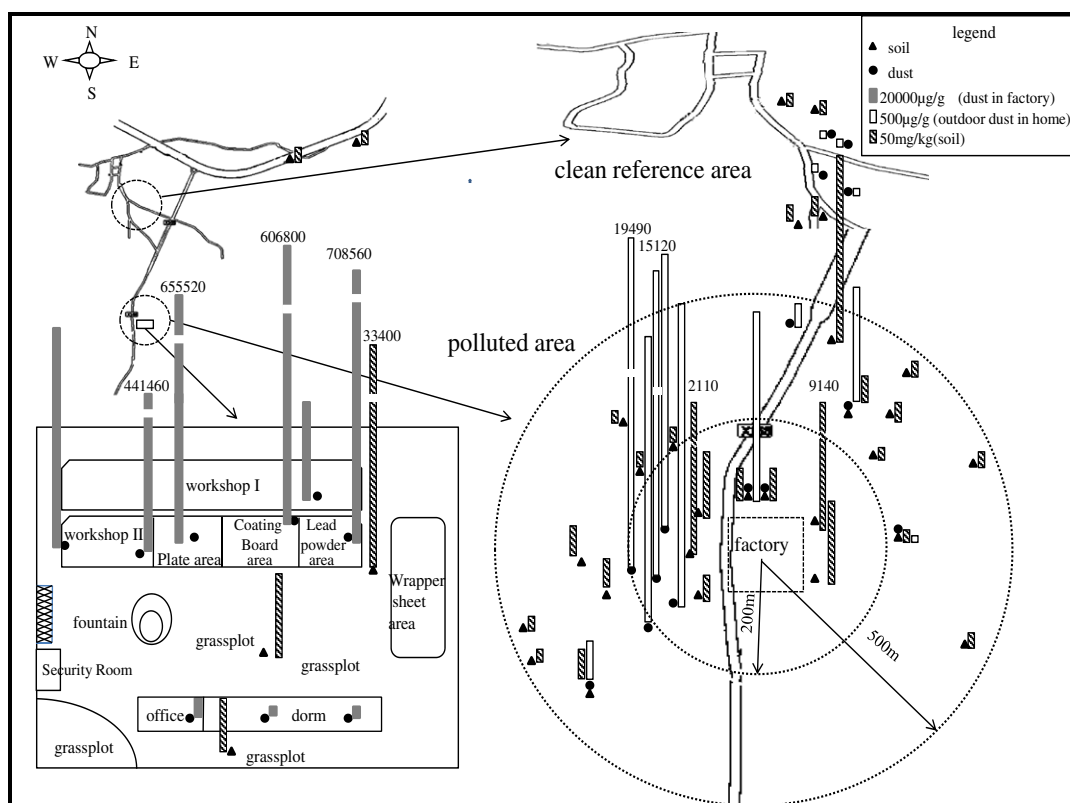


Fig. 1. Lead concentrations in soil and dust sampled around the factory.

0.085 µg for food samples, 0.30 µg for soil samples). As for the water blanks, the lead levels were below method detection limit (MDL). The recoveries in the spiked blanks were ranged from 90 to 115% (mean: 103%). The relative standard deviations (RSDs) for the food, soil and water samples were 0.51–18.9% (mean: 6.96%), 0.37–8.83% (mean: 3.60%) and 0.33–9.04% (mean: 3.29%), respectively. MDL in food, soil and water samples were 0.009–0.012 mg/kg, 4.94–9.87 mg/kg and 1.41 µg/L, respectively. Certified reference materials for soil (GSS-5) and rice (GSB-1) with certified values of 552 ± 44 mg/kg and 0.08 ± 0.03 mg/kg were purchased from National Research Center for Certified Reference Materials, China. They were analyzed with mean recoveries of 119% for GSS-5 ($n = 10$) and 98% of the certified value for GSB-1 ($n = 1$), respectively.

2.4. Statistical analysis

Due to the abnormal distribution of the data (Kolmogorov–Smirnov test, $p < 0.05$), all data was statistically processed using nonparametric tests (Kruskal–Wallis H and Mann–Whitney U), using SPSS v 13.0 for windows (SPSS Inc., Chicago, USA). The results of the statistical tests were considered statistically significant when $p < 0.05$. In those samples in which lead concentrations were lower than the MDL, half of the MDL was used. We excluded the possibility of contamination, because samples were collected without using metal tools or containers and analyzed under strict quality control and quality assurance. In addition, for those samples with unexpected high lead level, reanalysis was conducted.

3. Results and discussion

3.1. Characterization of human external exposure doses

3.1.1. Lead levels in the food

High lead concentration (4.36 mg/kg) was detected in a water convolvulus vegetable sample planted in the highly polluted soil

(2106 mg/kg) at a 40 m distance to the west of the factory. With the exception to this vegetable sample, lead levels in all other vegetable samples varied from 0.001 to 0.24 mg/kg with a mean value of 0.058 mg/kg. The average lead concentrations in vegetables were below allowable maximum levels of 0.3 mg/kg in foods, which are recommended by the Chinese Ministry of Health and National Standardization Administration (GB, 2762-2005).

The lead levels in the eggs and rice were 0.040–0.102 mg/kg and 0.048–0.137 mg/kg, with a mean value of 0.065 and 0.080 mg/kg, respectively. These levels were lower than the maximum allowed levels of contaminants in foods, which is 0.2 mg/kg, recommended by the Chinese Ministry of Health and National Standardization Administration (GB, 2762-2005).

3.1.2. Lead levels in the air

The average lead concentration in PM₁₀ in clean reference area ($0.14 \mu\text{g}/\text{m}^3$) was slightly lower than that of the polluted area ($0.18 \mu\text{g}/\text{m}^3$). These values were far below seasonal mean value ($1.5 \mu\text{g}/\text{m}^3$) and annual mean value ($1.0 \mu\text{g}/\text{m}^3$) provided by Chinese ambient air quality standard (GB3095-1996). A previous report for most European cities, the average lead levels in air were usually below $0.15 \mu\text{g}/\text{m}^3$ and 0.15 – $0.5 \mu\text{g}/\text{m}^3$ at non-urban and urban sites respectively (Delumyea and Kalivretenos, 1987). The lead levels in our study were slightly higher than those reported for in non-urban sites and lower than those in European cities (Delumyea and Kalivretenos, 1987). However, in 2007, the lead average levels in European countries reduced to $0.045 \mu\text{g}/\text{m}^3$ (Bierkens et al., 2011). In this study, since the air samples were collected after the factory production was suspended for a period of about 1 week, this data could not possibly reflect the real air lead pollution caused by the production activities.

3.1.3. Lead levels in the soil

Soil lead levels detected from 214 to 33,400 mg/kg (mean: 11,140 mg/kg) in the factory area, and found from 50.2 to 9140 mg/kg

(mean: 637 mg/kg) in the polluted area, and observed from 51.0 to 76.1 mg/kg (mean: 60.5 mg/kg) in the clean-reference area, respectively. The mean lead levels decreased in the order of: the plant area > the polluted area > the contrast area (Fig. 1). There were no significant differences between the factory, polluted and clean-reference areas ($p > 0.05$). In these three areas, soil lead levels decreased with the increasing distance from the central factory site. In the clean-reference area, soil lead levels ($n=6$) were in a narrower range with RSD of 15.0%, and this could represent the background soil lead level in the local area.

A higher level of lead (2106 mg/kg) was detected in one agricultural soil sample. However, in all other samples, lead levels were in the range of the secondary grade standard values (250–350 mg/kg) set for agricultural production, which was assigned by Chinese environmental soil quality standards (GB15618-1995). The average lead level in agriculture soils from the polluted area was 1.6 times higher than that from the clean reference area. In the agricultural soils from a zone at a 350 m distance from the plant site, lead levels (148.8 ± 104.2 mg/kg) were higher than 100 mg/kg, while the lead levels were 50–80 mg/kg in the soils from an area out in a 350 m distance from the plant site. As the distance increasing from the central factory, an obviously decreasing trend was observed for the soil lead levels.

3.1.4. Lead level in the water and sediment

Only lead level (1240 µg/L) in the production wastewater exceeded the upper limit value of 1000 µg/L, recommended by Chinese integrated wastewater discharge standard (GB 8978-1996). Lead levels in well water from both polluted and clean area varied from 0.71 to 9.35 µg/L with a mean value of 4.25 µg/L and no significant difference was observed in both areas ($p > 0.05$). Lead levels in tap water were from 0.71 to 4.59 µg/L with a mean value of 1.51 µg/L. The mean lead level in well water was 2.9 times higher than in the tap water. Both the lead levels in the well and tap water were below the value of 10 µg/L, recommended by the Chinese standards for drinking water quality (GB 5749-2006). In the surface water samples from ponds located in the polluted area and the clean reference area, Lead levels in both the polluted and clean area were below the first grade standard (10 µg/L) of the Chinese environmental quality standards for surface water (GB 3838-2002). The lead levels in sediments were 218.4 mg/kg and 67.8 mg/kg, respectively. Lead levels in both the water and sediment samples from the polluted area were higher than those from the clean reference area.

3.1.5. Lead levels in the indoor and outdoor dust

(1) Lead levels in the dust

Dust lead concentrations within the factory area varied from 441,460 to 708,560 mg/kg with a mean value of 601,030 mg/kg. Dust lead levels in the office and staff dormitories varied from 13,570 to 28,740 mg/kg with a mean value of 19,820 mg/kg. Lead levels from indoor dust samples collected in residential houses, were in a range of 506–10,950 mg/kg in the polluted area with the mean value of 5200 mg/kg, which were significantly higher than those in the clean reference area (234–610 mg/kg, mean: 331 mg/kg) ($p < 0.05$). In outdoor dust samples collected from work sites, the polluted area and the clean reference area, lead levels were 155,850–351,440 mg/kg (mean: 253,640 mg/kg), 270–19,490 mg/kg (mean: 9250 mg/kg), and 290–329 mg/kg (mean: 308 mg/kg), respectively. The lead levels in clean reference area varied in a small range (RSD = 5.5%), which was obviously lower than those in polluted area. This study assigned this lead level to represent a background levels in the outdoor dust in the clean reference area. If this assignment was reasonable, it meant that lead released from this battery factory should be responsible for the much higher lead levels found in the outdoor dust of the polluted area.

The lead levels in the dust samples declined with the increasing distances from the battery factory (Fig. 1). This suggested that this battery factory was a dominant lead emission source. Atmosphere dispersion of lead particles was a main venue which caused the neighboring environmental lead pollution problems. In order to understand whether the production of lead-acid battery emitted lead to the neighboring environment, we collected and analyzed particles from the production process and air pollution treatment facilities. The main process in the factory was to grind lead block for the lead-acid battery production. Lead powder and dust particles were detected by the laser particle size analyzer. The result showed that inhalable particle ($<10 \mu\text{m}$), fine particle ($<2.5 \mu\text{m}$) and ultrafine particle ($<1.0 \mu\text{m}$) covered 90%, 50% and 25% to lead powder (Fig. 2(a)), respectively. It indicated that lead powder would disperse into the surrounding environment if no dust remover was installed in the factory or the dust was removed by the filter incompletely. Particle size of the lead dust collected from cloth bag filter was also analyzed, in which inhalable particle and fine particle covered 32% and 18% (Fig. 2(b)), respectively. This indicated that the bag filter used in the technical process could not completely remove the ground lead powder, thus without preventing a considerable proportion (about 60%) of inhalable lead dust releasing to the environment.

Additionally, lead levels in outdoor dust samples around this battery factory decreased in varies directions in the order of: west and southwest > north > east (Fig. 1). The climate in the studied area is a typical subtropical monsoon climate. East wind is dominant from May to August per annum, while north wind is dominant in the other months (<http://www.cctcct.com>). This may explain the reasons why high lead concentrations in dust were found in the west and south-west of the battery factory during our study.

(2) Lead concentration ratio and relationship between outdoor and indoor dusts

In the polluted area, the lead level ratios of outdoor/indoor dust (O/I) of nine houses were greater than one and only one (H7) was less than 1 (Fig. 3). O/I ratios, greater than one, indicated that there existed a lead emission source in the circumjacent environment and that lead levels in indoor dust could be affected by the outdoor emission source. Except one house, with the highest lead level found in the outdoor dust, good correlation between outdoor and indoor dust lead levels was found ($R^2 = 0.756$, $p = 0.0023$), suggesting that outdoor lead emission source had affected indoor lead levels. In the clean reference area, O/I ratios were slightly higher than one in two houses, and less than one in the other two houses. In the adjacent kindergarten, the O/I ratio was less than one. The lead-containing toys and paints indoors may be the potential contributors for the observed lead levels, since it was reported that, in China, there were high lead levels in housing paints, and this exposure source was still not taken seriously (Lin et al., 2009) and remedied. However, this was outside of the focus of this study.

3.1.6. Lead levels in the tree leaves

Lead levels in the leaves of different kinds of tree varied from 36.6 to 440 mg/kg with a mean value of 164 mg/kg in the production area, from 0.26 to 46.0 mg/kg with a mean of 22.5 mg/kg in the polluted area, and from 0.14 to 4.73 mg/kg with a mean value of 2.87 mg/kg in the clean-reference area. As for the observed trends in soils and dusts, the lead levels in leaves tended to decrease from the plant area, to the polluted area, and further to the clean reference area. Fig. 4 showed that, other than grape tree leaves, lead levels in the leaves of the other tree decreased with the increasing distance from the plant location, and a decreasing trend was observed for the mango leaves lead levels with no significance ($p > 0.05$). The result indicated that this lead-acid battery factory have caused fairly higher lead levels in the plants planted its neighboring environment and factories.

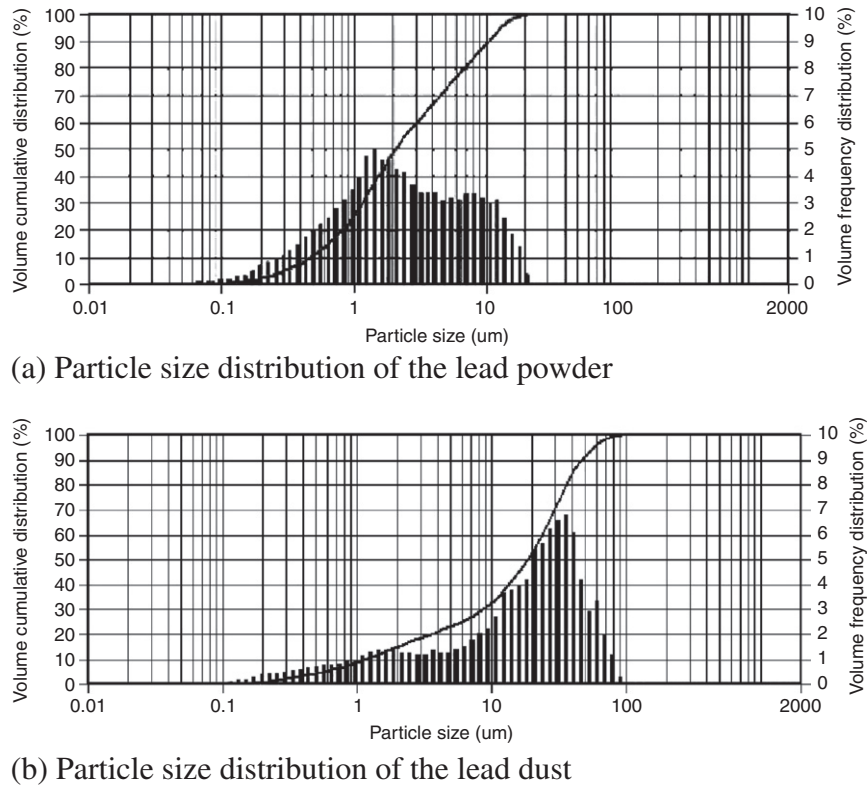


Fig. 2. (a) Particle size distribution of the lead powder.(b) Particle size distribution of the lead dust.

3.2. Calculating human exposure from external sources

Ingestion of the contaminated soil and dust, consumption of the contaminated food and drinking water, inhalation and dermal absorption of dust are main routes for the human lead exposure. Dermal absorption of inorganics is insignificant, though organic lead may be readily absorbed through the skin (U.S. ATSDR, 2005). Given that lead dust as inorganic, and emitted from the lead-acid battery factory, we would consider diet, inhalation and dust ingestion as the main exposure routes. The average daily doses (ADD) for lead were calculated using the equation from Supplemental material (Appendix A). Since only two air samples were collected, no conclusions could be drawn

about the inhalation exposure, therefore, the contribution of inhalation to daily lead intake was not included in this study. Compared to the contributions of diet and dust ingestion to daily lead intake, some studies had reported that the contribution from inhalation was negligible (Mielke and Reagan, 1998; Succop et al., 1998; Carrizales et al., 2006). The contributions of diet and dust ingestion to daily lead intake were calculated in this study (Fig. 5). In the clean reference area, primary lead exposure for adults was from dust ingestion (about 53%) and diet intake (about 47%). However, for toddlers, dust ingestion became the dominant route, contributing more than 70%. The contribution profile of different routes in the polluted area was obviously different compared with that in the clean

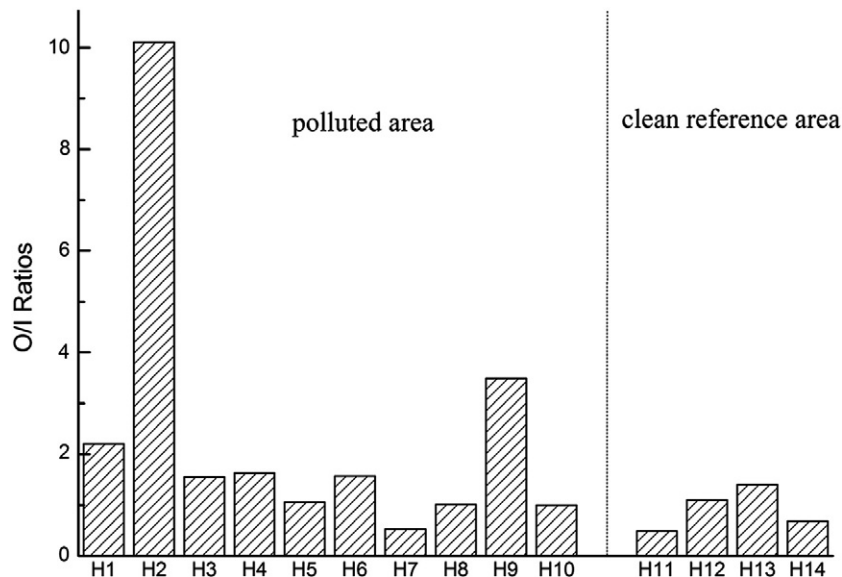


Fig. 3. The ratios of lead level between outdoor/indoor dust (O/I) from the polluted and clean reference areas.

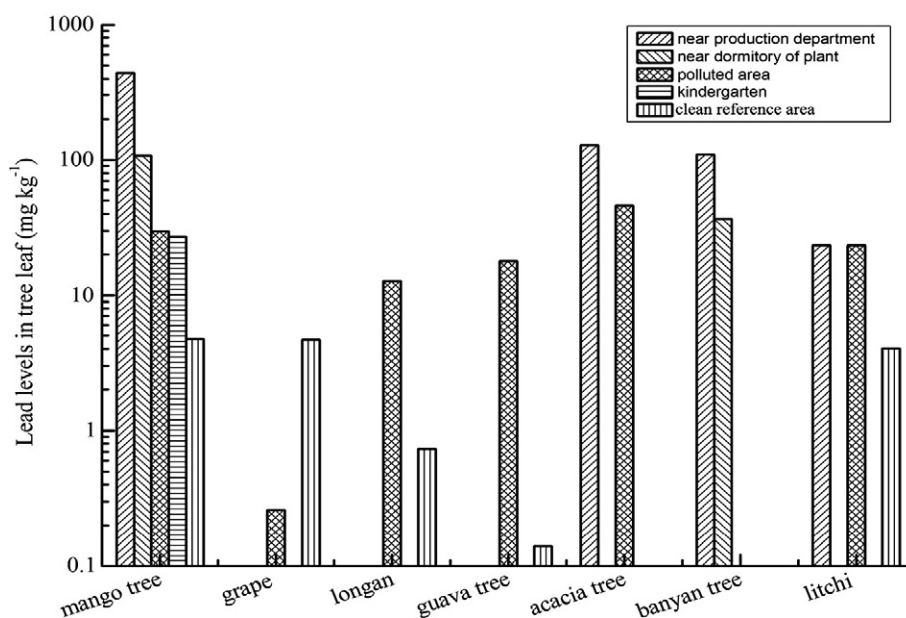


Fig. 4. Lead levels in different tree leaves from different studied areas.

reference area, where the dust ingestion became the dominant route and contributed over 90% to the total for both the adults and toddlers (Fig. 5). Several studies have reported positive correlations between heavy metal levels in indoor dust and children's high metal body burden, particularly for lead (Lanphear et al., 1996; Lanphera et al., 1998; Trepka et al., 1997; Meyer et al., 1999). The results of our study were comparable to the report on the children lead exposure near a copper-smelter in San Luis Potosi using the USEPA integrated exposure uptake biokinetic model (Carrizales et al., 2006).

3.3. Blood lead levels and risk assessment

3.3.1. Blood lead levels

Due to difficulty in collecting family members' blood samples, only blood samples from 11 families were collected from the polluted area and one family was collected from the clean reference area. The blood lead levels of the three-person family in the clean reference area were lower than 45 $\mu\text{g/L}$ (mean: 38.5 $\mu\text{g/L}$). This value is slightly higher

than the currently measured "baseline" blood lead above the limit levels of minimal anthropogenic origin (30 $\mu\text{g/L}$) (WHO, 2000). Of the 11 families living in the polluted area, blood lead levels for 17 adults and 15 children were available, and the lead levels for 16 adults and 13 children exceeded national standard value (100 $\mu\text{g/L}$) in China.

In this study, though the blood levels were observed in the limited families and members, and could not statistically reflect the blood lead levels of the residents living around the lead battery factory, we observed several basic characteristics. First, the blood lead levels in the clean reference area were significantly lower than those found in polluted area ($p < 0.05$). Second, the blood lead levels declined with the increasing distance from the factory. Third, the blood lead levels were positively related to the lead levels in dust (the average value of the sum of indoor and outdoor dust) (Fig. 6) ($R^2 = 0.379$, $p < 0.001$). The highest blood lead level value was 639 $\mu\text{g/L}$ in a family, corresponds to the environment they live in with the highest indoor dust lead level (12,440 mg/kg) detected

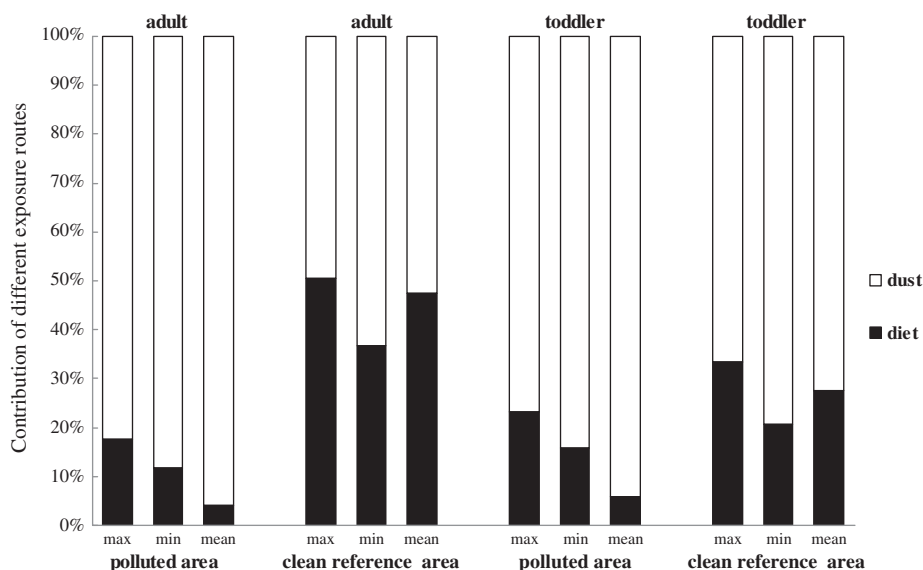


Fig. 5. Contribution of different exposure routes in the polluted and clean reference areas.

(Fig. 6). It suggested that dust ingestion may be the most important intake route for human exposure to lead in this environment in this study.

3.3.2. Risk assessment for lead exposure in the polluted and clean reference areas

Daily intakes of lead via the above-mentioned three routes were shown in Table 2. The mean lead exposure via diet for adult and toddler in the contrast area were comparable to those studies from Australia for children and adults (0.01–0.93 $\mu\text{g}/\text{kg}/\text{d}$) (Australia's submission to UNEP, 2005) and Finland for adults (0.24 $\mu\text{g}/\text{kg}/\text{d}$) (NFA, 2002 as cited by Finland's comments, 2006), but obviously lower than those from the studies for children and adults in USA (0.824–1.952 $\mu\text{g}/\text{kg}/\text{d}$) (U.S. ATSDR, 2005) and Mexico (3.5 $\mu\text{g}/\text{kg}/\text{d}$) (Mexico-comments, 2006). These values were also far lower than a provisional tolerable weekly intake (PTWI) of 25 $\mu\text{g}/\text{kg}$ of body weight (equivalent to 3.5 $\mu\text{g}/\text{kg}/\text{d}$) established by the joint FAO/WHO Expert Committee on Food Additives (UNEP, 2010). For adults, the total daily intake of lead in the polluted area was about ten times higher than that in the clean reference area. For toddlers, the total daily intake of lead within the polluted area was about sixteen times higher than that in the clean reference area.

For the adults and children in the clean reference area, the average and maximum total daily lead intakes were close to or lower than PTWI value of 3.5 $\mu\text{g}/\text{kg}/\text{d}$ (Table 2). However, for people living in the polluted area, the average total daily lead intakes were 6.19 and 50.1 $\mu\text{g}/\text{kg}/\text{d}$ for the adults and children, far higher than PTWI value, suggesting that living in the polluted area, the adults and children were at great risk for lead exposure. Young children (below 6 years old) are the most susceptible population to lead exposure, even at low levels of environmental pollution (UNEP, 2010). It may be inevitable that blood lead levels of the many children exceeded 100 $\mu\text{g}/\text{L}$ living in the polluted area. However, the average lead intakes for the people living in the polluted and clean reference area via the diet exposure route were far below PTWI value for the adults and children, which suggested that lead intakes via diet exposure routes were safe even in the polluted area.

Of the 17 adults, 4 had blood lead levels in the range of 300–400 $\mu\text{g}/\text{L}$, 7 in 150–250 $\mu\text{g}/\text{L}$ and one was less than 100 $\mu\text{g}/\text{L}$. When the lowest observed adverse effect levels (LOAELs) are 150–200 $\mu\text{g}/\text{L}$ and 200–300 $\mu\text{g}/\text{L}$, erythrocyte protoporphyrin elevation can occur for females and males. When LOAELs are as much as 300 $\mu\text{g}/\text{L}$ lead induced effects in adults are found, peripheral nerve dysfunction can appear (WHO, 2000). Of the 15 children, 2 had blood lead levels in the range of over 600 $\mu\text{g}/\text{L}$, 6 in

150–250 $\mu\text{g}/\text{L}$ and 5 in 100–150 $\mu\text{g}/\text{L}$. When LOAELs are 100–150 $\mu\text{g}/\text{L}$ and 150–200 $\mu\text{g}/\text{L}$, vitamin D3 reduction and cognitive impairment, erythrocyte protoporphyrin elevation may occur for the children. When LOAEL reach 700 $\mu\text{g}/\text{L}$, frank anemia can appear for the children (WHO, 2000). It is obvious that the adults and children in the polluted area are suffering from the potential lead poison. After the incident occurred, though the factory were suspended soon and local government had carried out timely treatment program for all the residents with blood lead levels exceeded Chinese national standard value (100 $\mu\text{g}/\text{L}$).

4. Conclusions

This study demonstrated that a considerable large proportion of inhalable and fine lead dust is generated from a lead acid factory entering the neighboring environments, even though bag filter is technically used in the production process in this factory. Atmospheric dispersion and deposition are the major transport pathways. The lead levels found in concentrations of soils and tree leaves in the polluted area were much higher than found in the clean reference area. However, lead levels found in circumjacent soil and underground water were lower than the recommended national standard values of China and no obvious lead pollution problem was observed in rice and vegetables growing in the polluted area. Ingestion of lead-contained dust was one of the major human exposure pathways, while the contribution of diet was relatively insignificant.

Our preliminary results from this study demonstrated that lead-acid battery production has caused severe environmental pollution problems and raised damaging public health issues. To our knowledge, there are about 2000 lead acid battery factories operating in China. Their environmental hazards still remain unknown. Due to the absence of health-related studies in these areas, urgent studies are needed to assess the adverse effects of lead exposure. Studies are being designed in our next step to further investigate the environmental and public health impacts from lead-acid battery factories. We expect that our results will become an important reference for environmental policy making and technological improvement for factory manufacturing processes.

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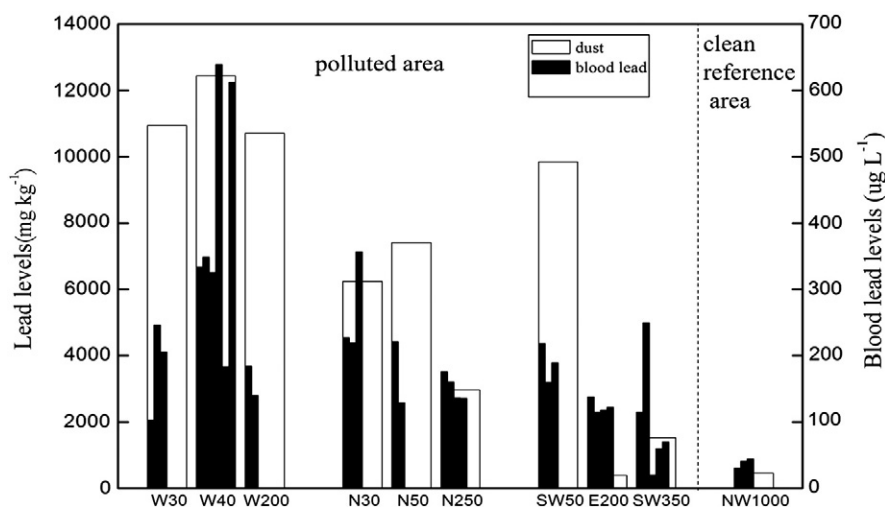


Fig. 6. The dust and blood lead levels in the human body studied. E, S, W and N represent the east, south, west and north of this plant, and the numeral represents the distance from the house to the studied factory.

Table 2Lead exposure levels in the polluted and clean reference areas for the studied adults and toddlers ($\mu\text{g/kg/d}$).

	Adult						Toddler					
	The polluted area			The clean reference area			The polluted area			The clean reference area		
	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
Σ diet	2.20	0.04	0.26	0.39	0.14	0.26	25.1	0.49	2.93	1.52	0.49	0.87
Dust ingestion	10.4	0.32	5.90	0.38	0.23	0.29	82.9	2.59	47.2	3.03	1.87	2.29
Σ total	12.6	0.36	6.16	0.77	0.37	0.55	108	3.08	50.1	4.55	2.36	3.16

Note: Σ diet = Σ vegetable + egg + rice + water), Σ total = Σ (diet + dust ingestion).

(zx_200809_16 and zx_200910_25). The authors thank Dr. Dongli Wang (Contamination Environmental Consulting, USA) for his reviewing the manuscript and giving many constructive comments and revisions.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2012.04.015>.

References

- Australia's submission to UNEP. *Pb in Australia*. Prepared by the Australian Government for the United Nations Environment Programme. 2005.
- Bierkens J, Smolders R, Holderbeke MV, Cornelis C. Predicting blood lead levels from current and past environmental data in Europe. *Sci Total Environ* 2011;409:5101–10.
- Carrizales L, Razo I, Téllez-Hernández JI, Torres-Nerio R, Torres A, Batres LE, et al. Exposure to arsenic and lead of children living near a copper-smelter in San Luis Potosi, Mexico: importance of soil contamination for exposure of children. *Environ Res* 2006;101:1–10.
- Chen L, Huang Y, Xu Z, Wen L, Peng X, Ye Z, et al. Human exposure to PBDEs via house dust ingestion in Guangzhou, south China. *Arch Environ Contam Toxicol* 2011;60:556–64.
- Chuang HY, Cheng WC, Chen CY, Yang YH, Sung FC, Yang CY, et al. A follow-up comparison of blood lead levels between foreign and native workers of battery manufacturing in Taiwan. *Sci Total Environ* 2008;394:52–6.
- Delumyea R, Kalivretenos A. Elemental carbon and lead content of fine particles from American and French cities of comparable size and industry, 1985. *Atmos Environ* 1987;21:1643–7.
- GB 11659-89. *Health protection zone standard for lead storage battery plants*. Ministry of Health of the People's Republic of China. (in Chinese)
- GB 15618-1995. *Environmental quality standard for soils*. Environmental Protection Administration of the People's Republic of China. (in Chinese)
- GB 2762-2005. *Maximum levels of contaminants in foods*. Ministry of Health of the People's Republic of China. (in Chinese)
- GB 3095-1996. *Ambient air quality standard*. State Environmental Protection Administration of the People's Republic of China. (in Chinese)
- GB 3838-2002. *Environmental quality standards for surface water*. State Environmental Protection Administration of the People's Republic of China. (in Chinese)
- GB 8978-1996. *Integrated wastewater discharge standard*. State Environmental Protection Administration of the People's Republic of China. (in Chinese)
- GB5749-2006. *Standards for drinking water quality*. Ministry of Health of the People's Republic of China. (in Chinese)
- Gulson BL, Davis JJ, Mizon KJ, Korsch MJ, Law AJ, Howard D. Lead bioavailability in the environment of children: blood lead levels in children can be elevated in a mining community. *Arch Environ Health* 1994;49:326–31.
- Hsiao CY, Wu HDI, Lai JS, Kuo HW. A longitudinal study of the effects of long-term exposure to lead among lead battery factory workers in Taiwan (1989–1999). *Sci Total Environ* 2001;279:151–8.
- <http://news.cntv.cn/20110603/110006.shtml> [cited 2011 Agu 30].
- http://news.xinhuanet.com/politics/2011-05/17/c_121423661_2.htm [cited 2011 Agu 30].
- <http://www.cctct.com/travel-guide/1-20/1877/51769.html> [cited 2011 Agu 30].
- Huang Y, Chen L, Peng X, Xu Z, Ye Z. PBDEs in indoor dust in South-Central China: characteristics and implications. *Chemosphere* 2010;78:169–74.
- IPCS. Inorganic lead—Environmental Health Criteria 165. International Programme on Chemical Safety. Geneva, Switzerland: World Health Organisation; 1995.
- Ji A, Wang F, Luo W, Yang R, Chen J, Cai T. Lead poisoning in China: a nightmare from industrialisation. *Lancet* 2011;377:1474–6.
- Lanphear BP, Weitzman M, Winter NL, Eberly S, Yakir B, Tanner M, et al. Lead-contaminated house dust and urban children's blood lead levels. *Am J Public Health* 1996;86:1416–21.
- Lanphear BP, Matte TD, Rogers J, Clickner RP, Dietz B, Bornschein RL, et al. The contribution of lead-contaminated house dust and residential soil to children's blood lead levels. *Environ Res* 1998;79:51–68.
- Lin GZ, Peng RF, Chen Q, Wu ZG, Du L. Lead in housing paints: an exposure source still not taken serious for children lead poisoning in China. *Environ Res* 2009;109:1–5.
- Mexico-comments. Comments to the draft report from Comisión Federal para la Protección contra Riesgos Sanitarios, Secretaría de Salud de México; 2006.
- Meyer I, Heinrich J, Lippold U. Factors affecting lead, cadmium, and arsenic levels in house dust in a smelter town in eastern Germany. *Environ Res* 1999;81:32–44.
- Meyer PA, Brown MJ, Falk H. Global approach to reducing lead exposure and poisoning. *Mutat Res* 2008;659:166–75.
- Mielke HW, Reagan PL. Soil is an important pathway of human lead exposure. *Environ Health Perspect* 1998;106(Supplement 1):217–29.
- NFA. Riskraportti: Elintarvikkeiden ja talousveden kemialliset vaarat (Risk report: chemical hazards of food and drinking water). Helsinki: National Food Agency; 2002. valvontaopas-sarja 2/2002 (as reported by Finland Comments, 2006).
- Paoliello MMB, Capitani EMD, Cunha FGD, Matsuo T, Carvalho MDF, Sakuma A, et al. Exposure of children to lead and cadmium from a mining area of Brazil. *Environ Res Sec A* 2002;88:120–8.
- Succop P, Bornschein R, Brown K, Tseng CY. An empirical comparison of lead exposure pathway models. *Environ Health Perspect* 1998;106(Supplement 6):1577–83.
- Trepka MJ, Heinrich J, Krause C, Schultz C, Lippold U, Meyer E, et al. The internal burden of lead among children in a smelter town. *Environ Res* 1997;72:118–30.
- U.S. ATSDR. *Toxicological profile for lead*. (Draft for public comment). U.S. Department of health and human services. Public Health Service. Agency for toxic substances and Disease Registry, Atlanta, U.S.A. 2005.
- UNEP. Final review of scientific information on lead. United Nations Environment Programme Chemicals Branch, DTIE; 2010.
- VDI (German Association of Engineers). Measurement of indoor air pollution—sampling of house dust, VDI 4300 Part 8. Berlin: Beuth-Verlag; 2001.
- WHO. Air quality guidelines for Europe. WHO Regional Publications, European Series, No. 91; 2000.
- Zhao RR, Ren AF, Chen HY. Current problem and perspectives of lead-acid battery industry in China. *Battery Bimon* 2009;39:333–4. (in Chinese).